

# Anomalous high energy dispersion in photoemission spectra from insulating cuprates

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Angle resolved photoelectron spectroscopic measurements have been performed on an insulating cuprate  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ . High resolution data taken along the  $\Gamma$  to  $(\pi, \pi)$  cut show an additional dispersive feature that merges with the known dispersion of the lowest binding energy feature, which follows the usual strongly renormalized dispersion of  $\approx 0.35$  eV. This higher energy part reveals a dispersion that is very close to the unrenormalized band predicted by band theory. A transfer of spectral weight from the low energy feature to the high energy feature is observed as the  $\Gamma$  point is approached. By comparing with theoretical calculations the high energy feature observed here demonstrates that the incoherent portion of the spectral function has significant structure in momentum space due to the presence of various energy scales.

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Since the early days of high temperature superconductivity (HTSC), it has been suggested, based on the cuprate phase diagram, that the microscopic origin of HTSC may hinge on how charges move in a background of strong antiferromagnetic (AF) interactions. Thus, a natural starting place is to learn how a single hole behaves in a half-filled AF system. This is precisely what is measured by angle resolved photoelectron spectroscopy (ARPES) on the half-filled cuprates. Indeed, such considerations have motivated many ARPES studies on parent HTSC compounds such as  $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ ,<sup>1,2,3,4</sup>  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ ,<sup>5</sup>  $\text{Nd}_2\text{CuO}_4$ ,<sup>6</sup> and lightly doped  $\text{La}_2\text{CuO}_4$ .<sup>7</sup>

While Local Density Approximation (LDA) calculations predict these systems to be metallic with the width of the lowest filled energy band  $\sim 2$  eV,<sup>8</sup> optical measurements indicate they are insulators with a band gap of  $\sim 1.5$  eV,<sup>9,10</sup> which results from the strong on-site Coulomb repulsion. ARPES results have revealed many other important features of insulating cuprate compounds. Energy Distribution Curve (EDC) analysis has revealed a broad low-energy charge transfer band feature,<sup>1</sup> which is commonly referred to as a Zhang-Rice singlet. It has a band width of 0.35 eV which has been related to a renormalization by the exchange interaction  $J$ . The dispersion has been well described by the  $t$ - $t'$ - $t''$ - $J$  model.<sup>2</sup> In addition, results from  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$  show a  $d$ -wave-like dispersion and a feature in the occupation probability  $n(k)$  that was termed as a ‘remnant’ Fermi surface.<sup>5</sup>

All these results were obtained with a lower angular (that is, momentum) resolution of  $2^\circ$  compared to what is commonly used now ( $0.25^\circ$ ), yet made important contributions in understanding hole motion in the AF background. As has been found across the phase diagram of the cuprates, the dramatic increase in photoemission resolution has revealed new features in the data which were previously unresolved.<sup>11</sup> In this paper, we present high

$k$ -resolution data from  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$  which reveal yet another surprising result. The lowest energy excitations near  $(\pi/2, \pi/2)$  remain consistent with the above description, but at higher binding energies we observe a broad rapidly dispersing feature which tracks the unrenormalized band dispersion. We note that there is significant transfer of spectral weight from the low energy feature to the high energy one as the  $\Gamma$  point is approached. This description is consistent with several *different* numerical calculations,<sup>18,19,20,21,22</sup> and our data experimentally confirms this high energy electronic structure. We will discuss our observations in the context of the current theoretical understanding.

$\text{Ca}_2\text{CuO}_2\text{Cl}_2$  single crystals were grown by a flux method.<sup>12</sup> Experiments were performed at beamline V of the Stanford Synchrotron Radiation Laboratory (SSRL). Samples, which were oriented *ex situ* prior to the experiments by Laue x-ray diffraction were cleaved *in situ* at a base pressure better than  $5 \times 10^{-11}$  torr. Spectra were taken with 25.2, 22.4, and 15.5 eV photons, the total energy and angular resolution was typically 30 meV and  $0.25^\circ$ , respectively, and no sign of charging was observed in these samples.

Figure 1(a) shows EDCs of the lowest energy feature along the nodal direction (that is,  $k_x = k_y$ ) from tetragonal  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ . It is seen that a broad peak develops near  $(\pi/4, \pi/4)$ , gains spectral weight, and disperses toward the lower binding energy side as  $k$  increases from  $\Gamma$  to  $(\pi/2, \pi/2)$ . It then suddenly loses intensity near  $(\pi/2, \pi/2)$ . Furthermore, it leaves no spectral intensity at the Fermi energy  $E_F$ , as expected for an insulator. The total dispersion of the peak is about 0.35 eV. Note also that there is very little spectral intensity near the  $\Gamma$  point. These are the salient features of the insulating parent HTSC compounds and have been discussed in the earlier literature.<sup>11</sup>

Our high  $k$ -resolution data, however, reveal a feature

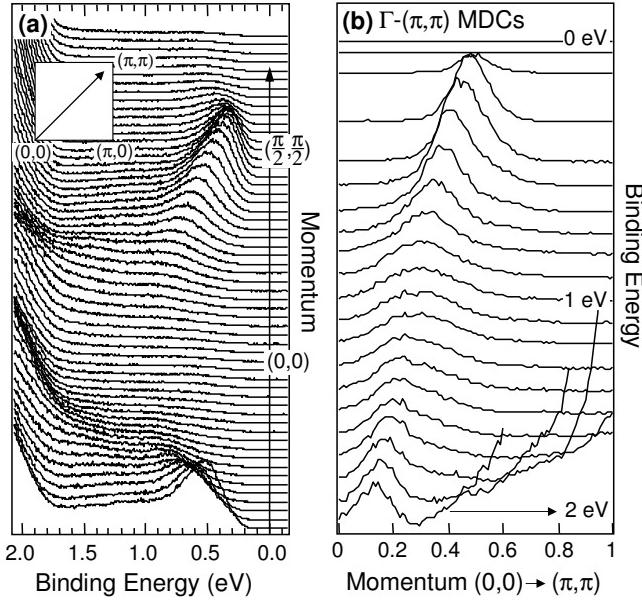


FIG. 1: ARPES data from  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$  along the  $\Gamma-(\pi,\pi)$  direction, with 25.2 eV photons. (a) EDC's with the  $k$  point for each curve spaced evenly along the  $\Gamma-(\pi,\pi)$  line. (b) MDC curves of the ARPES data shown at equal binding energy intervals, from 2 eV (bottom) to the chemical potential (top). The large spectral weight near  $(\pi,\pi)$  for high binding energy MDCs is due to the  $\text{O}_{2p_\pi}$  states in the main valence band.

that was not previously observed. The details can be seen better with an intensity plot. The data in figure 1 are symmetrized about the  $\Gamma$  point and plotted in Figure 2. The low energy feature described above is clearly seen between  $k = (\pi/4, \pi/4)$  and  $(\pi/2, \pi/2)$  with total dispersion of about 0.35 eV. Examining the image, one can clearly see that there is a fast dispersing feature above 0.8 eV binding energy, which is in sharp contrast with the known and slowly dispersing low energy feature. An indication of this additional feature was also seen in the earlier low resolution data roughly 500 meV below the lowest energy feature whose origin was uncertain, but was attributed to a string resonance excitation.<sup>2</sup> Even though not as clear as in the  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$  case, we also observed a similar high energy feature in  $\text{Sr}_2\text{CuO}_2\text{Cl}_2$  (not shown).

Due to the rapid dispersion this feature is most systematically characterized by the Momentum Distribution Curves (MDCs) shown in figure 1(b). The MDC curves are well fit by a Lorentzian up to binding energies of 2 eV at which point the main valence band dominates the spectral weight. The resulting peak positions determined by fitting the MDCs are overlaid on figure 2 from which both the low and high energy dispersions discussed above are evident. One can also track the dispersion from the peak positions of the EDCs in figure 1(a) which are marked with circles and triangles. The dispersion obtained from the MDCs and EDCs between 0.3 and 0.7 eV (low energy part) roughly agree. At high energies ( $\sim$

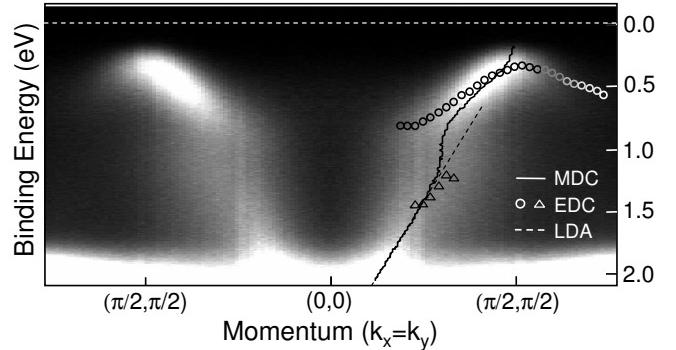


FIG. 2: Intensity plot of data shown in figure 1 as functions of the binding energy and momentum. The data was symmetrized around the  $\Gamma$  point. Also shown on the plot are the dispersions obtained by following the peak positions of the MDCs (solid line) and the EDCs (circles and triangles). The results are compared with the shifted dispersion from the LDA calculation (dashed line).

1.3 eV) the MDCs also agree with the very broad and weak feature seen in the EDCs (triangles). The fact that the MDCs contain a single peak at all energies suggests that the suppressed spectral weight near the  $\Gamma$  point is not caused by the matrix element effect, but is due to a significant transfer of spectral weight from the low energy feature to the rapidly dispersing high energy feature. Also plotted in the figure is the dispersion from the LDA calculation shifted by  $\sim 0.7$  eV (dashed line). Even though there is no independent justification for shifting the LDA dispersion by 0.7 eV, the agreement between the slopes of the two dispersions is still remarkable. This suggests that the rapidly dispersing high energy feature may be tracking the unrenormalized band.

The low and high energy dispersive features form a kinklike feature as can be seen in the image of figure 2. Ignoring the energy scale, this is very reminiscent of the data on metallic hole-doped cuprates, where a kink in the quasi-particle dispersion has been attributed to coupling to a phonon mode of  $\sim 70$  meV.<sup>13</sup> In our data the kinklike feature occurs roughly 0.5 eV below the top of the valence band, which can be seen from the MDC derived dispersion in figure 2. However, in the present case the kink can not originate from coupling to a bosonic mode. First of all, the system under study is an insulator with a 1.5 eV charge transfer gap and coupling to a bosonic mode is expected to happen only in metallic systems in the standard theory. In addition, we also stress that, from the extremely large widths of the EDC's and MDC's, the features we are studying are certainly not quasi-particles.<sup>14</sup> In regards to the latter point, the MDC width of the high energy feature clearly *decreases* as the binding energy increases above 1 eV (see figure 1(b)). This is counterintuitive as the phase space for decay channels of the photohole should increase as the binding energy increases, which should thus further raise a red flag when attempting to interpret these features as

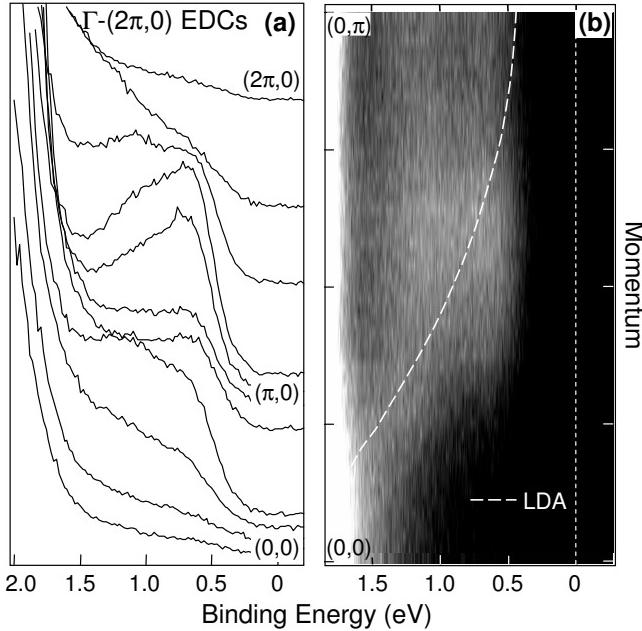


FIG. 3: (a) EDCs along the  $\Gamma$ - $(2\pi, 0)$  direction taken with 22.4 eV photons. The spectral weight at  $(2\pi, 0)$  is weak, as is the case for  $\Gamma$ . (b) Image plot of ARPES data along the  $\Gamma$ - $(0, \pi)$  cut with 15.5 eV photons. The in-plane polarization of the light is along the  $(\pi, 0)$  direction. Overlaid on the plot is the dispersion from LDA calculation shifted by 0.2 eV (dashed line)

quasi-particle bands.

The apparent transfer of spectral weight between the two observed dispersing features may give an answer to the puzzling observation of the vanishing spectral weight at the  $\Gamma$  point in all cuprates. Even though the dispersions in cuprates, especially in insulating compounds, have been said to be renormalized to one with  $\sim 0.3$  eV dispersion, the charge transfer peak mysteriously disappears near the  $\Gamma$  point. Traditionally, this lack of spectral weight at  $\Gamma$  has been attributed to matrix element effects.<sup>5</sup> The photoemission intensity is roughly expressed as  $I \sim |\langle i | \mathbf{A} \cdot \mathbf{p} | f \rangle|^2$  where  $|i\rangle$  and  $|f\rangle$  are initial and final states, respectively. Both the initial  $|i\rangle$  (primarily Cu  $3d_{x^2-y^2}$ ) and final states  $|f\rangle$  ( $\approx e^{ikz}$ ) have even symmetry with respect to the  $(\pi, 0)$  direction at the  $\Gamma$  point. Since  $\mathbf{A} \cdot \mathbf{p}$  is a dipole operator, it provides an odd symmetry if the polarization is in the plane and along the  $(\pi, 0)$  direction. This has been the argument for the vanishing spectral weight at the  $\Gamma$  point. In real cases, however, such symmetry condition is never perfectly met, as the polarization almost always has an out-of-plane component (taken here as the  $z$ -direction).<sup>15</sup> This additional  $z$ -component contributes to a non-vanishing matrix element, and thus the spectral weight should be finite at the  $\Gamma$  point. In fact, the manganites do possess significant spectral weight at  $\Gamma$  due to a band with  $d_{3z^2-r^2}$  orbital character for which the above arguments would be identical.<sup>16</sup>

Perhaps even more importantly, we have failed to find any photoemission intensity at the equivalent  $k$ -space location of  $(2\pi, 0)$  where the selection rule is no longer relevant. This can be seen in figure 3(a) where upon approaching  $(2\pi, 0)$  from  $(0, 0)$  spectral weight is rapidly pushed to higher binding energies, and the spectra at  $(2\pi, 0)$  look identical to  $(0, 0)$ . Furthermore, note that the matrix element argument dictates that the entire  $(0, 0)$  to  $(0, \pi)$  cut should have vanishing weight due to the fact that the in-plane component of polarization of the light is along the  $(\pi, 0)$  direction. Although it is indeed suppressed with respect to the  $(0, 0)$  to  $(\pi, 0)$  cut, there still remains finite low energy weight above  $(0, \pi/4)$ , as can be seen in figure 3(b). This not only vanishes upon approaching the  $\Gamma$  point, but also has a high binding energy feature which disperses rapidly to higher binding energy. This is identical to the  $\Gamma$  to  $(\pi, \pi)$  cut, including the observation that the rapidly dispersing high energy feature has a similar dispersion to that of LDA, as can be seen in panel 3(b).

All of this suggests that in addition to the matrix elements which will suppress spectral intensity at the  $\Gamma$  point, there is a transfer of spectral weight to very high binding energies as the  $\Gamma$  point is approached. This is a necessary consequence of the ARPES sum rules which say that the total spectral weight of the single particle spectral function must be conserved ( $\int A(k, \omega) d\omega = 1$ ). Note, this statement is true even for highly  $k$  dependent self energies.

Theoretically, there are several potential explanations for the multiple features in the photoemission spectra. Cluster calculations identify the first hole ionization state as a singlet state and also calculate a large spectrum of excitations which one will observe in a photoemission experiment.<sup>17</sup> While the majority of the states lie too high in energy (such as the Zhang-Rice triplet state at  $\sim 3.5$  eV), there are states which are only  $\sim 1.5$  eV above the lowest lying singlet state. Although we can not rule this out conclusively, these states are still somewhat too large in energy, and there is also no *a priori* reason to expect these states to mimic the dispersion of the underlying band structure. Thus it is unlikely that they should correspond to our rapidly dispersing band-like feature. Alternatively, momentum resolved numerical calculations of the single band Hubbard model and the extended t-J model have large amounts of incoherent spectral weight (>50% at many  $k$ -points) with significant structure in momentum space spread over large energies. Surprisingly, although approached differently from the numerical algorithm and approximations chosen, these calculated results bear similarities to what we observe in our data, whether it be from exact diagonalization,<sup>18</sup> self-consistent Born approximation,<sup>19</sup> quantum Monte Carlo,<sup>20</sup> strong coupling perturbation theory,<sup>21</sup> or variational cluster perturbation theory<sup>22</sup> (a type of generalized dynamical mean field theory). We also note that the high energy structure was sometimes referred to as "string" states in the earlier literature.<sup>23</sup> The recent calculations

show as  $\Gamma$  is approached spectral weight is transferred from a low energy band that mimics that of the  $t$ - $J$  model to spectral weight at higher energies with an energy scale of the hopping energy  $t$ , similar to what we see experimentally. The two features have been interpreted in terms of the coherent motion of a hole in the antiferromagnetic spin background and the incoherent motion of a hole hopping within the antiferromagnetic spin bag.<sup>20</sup> Thus, one might naturally expect the incoherent feature to track the unrenormalized band.

The outstanding problem with most of the numerical calculations is that the lowest energy excitation still corresponds to some sort of quasiparticle like pole, which is not consistent with ARPES experiments. The very large widths both in EDCs and MDCs are unphysical lifetimes for an associated quasiparticle. Also, the low-energy portion of the EDC lineshape can not be fit by any sort of power-law tail as would be expected from a quasiparticle picture.<sup>24</sup> Finally, the doping dependence of the spectral weight argues that the quasi-particle states which are formed at the chemical potential in the hole-doped compounds receive spectral weight from the incoherent background which is the same feature we identify as the charge transfer band in the insulator.<sup>24,25</sup>

A fully consistent picture of the insulator must therefore also be able to explain the large widths which are observed. An obvious candidate for the cause of such large widths is the strong electron-electron correlation, which suggests that a multiple state picture be naturally evoked in the photoemission spectra and that the true quasiparticle has vanishing weight.<sup>14</sup> In support of such a view, the phase string calculations based on the  $t$ - $J$  model have a vanishing quasi-particle residue, and the spectra, composed entirely of incoherent weight, reasonably reproduce the experimental lineshapes and dispersion.<sup>26</sup> Alternatively, the multiple states that constitute the broad peaks observed in the experiments may be many shake-off peaks as in the case of the Franck-Condon effect.<sup>24</sup> The appealing aspect of these types of pictures is that, as we have already demonstrated, our highly dispersing feature in  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$  can most naturally be explained as being structure in the incoherent part of the spectral function. Indeed, in support of the latter view some calculations

predict that the broad peaks still track the  $t$ - $J$  dispersion even when the polarons are of lattice origin.<sup>28,29</sup> The large widths of the insulator can then be explained as resulting entirely from the structure in the incoherent spectral function which tracks both energy scales in the problem: a ' $J$ '-like band at low energy and a ' $t$ '-like band at high energies.

In conclusion, we use high resolution ARPES data of  $\text{Ca}_2\text{CuO}_2\text{Cl}_2$  to elucidate a rapidly dispersing feature at high binding energies, which is very close to the unrenormalized bare band. We find a transfer of spectral weight as a function of momentum from the low energy feature, which tracks the extended  $t$ - $J$  model dispersion, to this high energy feature as  $\Gamma$  is approached. This interpretation is consistent with several different calculations working on the half-filled  $\text{CuO}_2$  plane. This effect is in addition to the matrix element effects which we believe are insufficient to explain the lack of spectral weight at  $\Gamma$ . The high-energy feature highlights the fact that the incoherent spectral weight is spread over very large energies and has significant structure in momentum space. This potentially allows a previously overlooked aspect of the spectral function to be used to differentiate various models which purport to understand the spectra of strongly correlated electron systems.

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